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Midinfrared emission from InGaN/GaN-based light-emitting diodes

Daniel Hofstetter[a] and Jérôme Faist
Institute of Physics, University of Neuchâtel, Neuchâtel, CH 2000, Switzerland

David P. Bour
XEROX Palo Alto Research Center, Palo Alto, California 94304

Midinfrared emission on violet, blue, and green InGaN light-emitting diodes has been measured between 85 and 300 K for various injection current densities. We found that the diode with the highest In composition in the active region had the shortest midinfrared emission wavelength and vice versa. With increasing In content, a significantly decreasing amount of TM polarization was observed in the midinfrared emission spectrum. This result suggests that the density of states in the higher-In content devices corresponds to a zero-dimensional electronic system rather than a two-dimensional electron gas. In contrast to this, the violet light-emitting diode exhibited a higher degree of TM polarization; similar to a red InGaP-based quantum-well device.

In recent years, there has been an increasing interest in midinfrared (mid-IR) quantum-cascade (QC) light sources, especially for applications in environmental sensors.[1,2] Up to this point, QC lasers fabricated from the InGaAs/InAlAs and GaAs/AlGaAs material systems have been demonstrated.[3,4] The InGaAs/InAlAs-based devices have already reached a high level of maturity and are commercially available. Nevertheless, there is an ongoing interest for the identification and implementation of new material systems other than the ones mentioned above. More particularly, there exists, so far, no QC light source with an emission wavelength shorter than 3.4 μm. This shortest wavelength was achieved with strain-compensated InGaAs/InAlAs, which has a conduction-band discontinuity of 740 meV.[5] For the III nitrides, the combination of GaN and AlGaN would offer already more than 1 eV conduction-band discontinuity. For these reasons, several research groups are looking into the issue whether the III nitrides could be successfully implemented as a base material for shorter-wavelength QC light emitters.

For a mid-IR intersubband transition taking place in a quantum well (QW), one expects, due to the polarization selection rule, a strongly TM-polarized edge emission. On the other hand, a device with a bulk active region would show no such polarization effect. Moreover, because of the rectangular shape of an ideal QW, only weak band filling should occur at low-injection-current densities. As two typical, well-understood examples, we investigated the mid-IR emission of red InGaP-based LEDs with both a QW and a bulk active region. The former device was strongly TM polarized and showed almost no band filling, while the latter was basically unpolarized. Similarly, our mid-IR luminescence experiments on InGaN-based material did not clearly reveal the TM-polarization selection; neither did we see the expected weak band filling. According to Refs. 6 and 7, InGaN with high-In mole fraction tends to phase separate already during growth. This phenomena has been described by several authors using transmission electron microscopy, x-ray diffraction, absorption measurements, and also luminescence studies.[6–8] In order to confirm a dependence on the In mole fraction in the active region, we will compare nitride LEDs with different In compositions.[9,10]

The LEDs we investigated have an InGaN-based active region and emit violet ($\lambda = 400$ nm), blue ($\lambda = 475$ nm), and green ($\lambda = 525$ nm) light. For comparison, we studied also the behavior of a red InGaP ($\lambda = 670$ nm) LED. The visible emission spectra of these samples have been measured using a Jobin–Yvon grating spectrometer with a focal distance of 0.4 m. The violet, blue, and green LEDs were pumped at 80, 160, and 240 mA and the red one at 20, 40, and 60 mA. Taking into account the different active areas of the four LEDs, this corresponds to maximal current densities of 1.0 kA/cm^2 (violet), 0.6 kA/cm^2 (blue/green), and 1.2 kA/cm^2 (red), respectively. The emission spectra of all LEDs are reported in Fig. 1. For the blue/green InGaN-based LEDs, we see a substantial shift towards higher energies when driving the devices at higher-injection currents, whereas the violet and the red LEDs exhibit a small wavelength shift towards

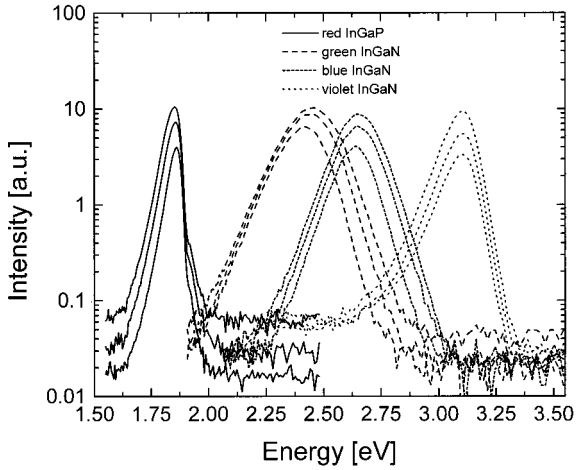

FIG. 1. Visible emission spectra of the interband electroluminescence measured on violet, blue, and green InGaN-based, and red InGaP-based LEDs.

[a]Electronic mail: Daniel.Hofstetter@iph.unine.ch

lower energies. The high-energy shift of the blue and green LEDs is essentially understood as a band-filling effect. If the active region were an ideal QW, then the density of states would be constant and the effect should be much weaker. A certain effect of the internal piezoelectric field, which is screened under high carrier injection, cannot be excluded; however, this effect is usually rather small for narrow QWs.[11] The redshift of the luminescence peak in the red and violet LEDs can be explained by an almost complete absence of band filling and the dominance of band-gap shrinkage. The difference in the spectral width of the red and the violet/blue/green devices is also an indication for the different density of states in the two sorts of active regions. Since the bands of the red InGaP and the violet InGaN LEDs do not fill quickly, there will be mainly band-edge to band-edge recombination, which results in a narrow energy range of the luminescence. On the other hand, the more quantum-box-like density of states of the blue/green InGaN LEDs leads to rapid band filling, and thus to a much larger variety of possible transition energies. The InGaN/GaN material system itself shows an increasing tendency to phase separate when increasing the In content;[12] therefore, the quite considerable difference in spectral width between violet, blue, and green LEDs can be understood on the base of this argumentation as well.

According to Ref. 13, one can measure the carrier temperature of a two-dimensional electron gas in a QW by determining the slope of the exponential decay on the high-energy tail of the visible emission peak. Since the active region of the blue/green InGaN LEDs forms not an ideal QW, their high-energy slopes on the luminescence peak correspond, as expected, to a too high electronic temperature of 550 K. Similarly, if one tries to measure the carrier temperature on the electroluminescence data of O'Donnell, Martin, and Middleton,[8] one finds a carrier temperature of 110 K. The reason for the inaccurate carrier temperature measurement is the modified (not constant) density of states in the highly phase-separated active material of the blue/green LEDs. In contrast to this, the violet/red LEDs exhibit a carrier temperature of 330 K, which is only about 20 K higher than the estimated value of the lattice temperature. Evidently, violet/red LEDs show a QW-like behavior, whereas the active region of blue/green LEDs seems to consist of a more quantum-box-like material.

In order to successfully measure a mid-IR emission from the commercial blue/green devices, it was necessary to remove their plastic caps. The devices were then placed into a temperature-controlled N_2 flow cryostat. The light from the edge was collected by $f/0.8$ optics and fed into a high-resolution Fourier transform spectrometer (Nicolet Magna-IR 860), where we detected it using a liquid-nitrogen-cooled HgCdTe detector. For these experiments, we used current pulses of 5 μs, and a pulse repetition frequency of 100 kHz.

Edge emission mid-IR luminescence spectra at 85 and 300 K for violet, blue, green, and red LEDs are shown in Fig. 2. At room temperature, we observed peak energies of 1650, 1800, and 1950 cm^{-1} for the violet, blue, and green LEDs, respectively. As for the visible emission, there is again a correlation between the band gap and transition energy. But this time, the situation is inverse to what was described above. A higher-In content of the active region leads to deeper QWs, and therefore, to larger transition energies, as illustrated by the insets of Fig. 2. Taking into account that, for these devices, a QW is a rather poor approximation, good agreement was found between the measured emission peaks and the calculated transition energies. The calculated emission wavelengths for transitions between the first excited state and the electronic ground state were 1720, 2000, 2180 cm^{-1} for the violet, blue, and green LEDs, respectively.

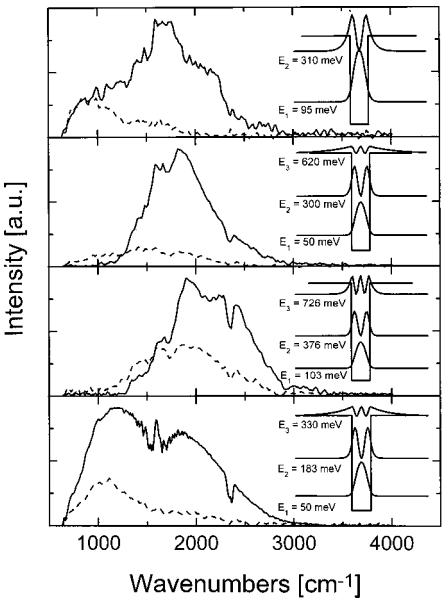

FIG. 2. Midinfrared intersubband luminescence spectra of violet, blue, green, and red LEDs at 85 and at 300 K. The inset show the QWs with calculated wave functions for each device. We used $d=30$ Å, $\Delta E_c=375$ meV for the violet; $d=35$ Å, $\Delta E_c=620$ meV for the blue; $d=35$ Å, $\Delta E_c=820$ meV for the green; and $d=65$ Å, $\Delta E_c=335$ meV for the red LED.

At 85 K, the emission of all devices became much weaker and was partially masked by background blackbody radiation. A similar decrease of population efficiency, and thus intersubband luminescence intensity at lower device temperature, has been described on optically pumped GaAs QWs.[14] This observation confirms that the excited states of our LEDs are populated mainly by band filling combined with electron heating. In fact, the high-injection voltage of almost 8 V at 240 mA will certainly result in electron heating, as indicated by the carrier temperature measurement on the violet LED. Furthermore, because of the interband recombination lifetime being much longer than the intersubband lifetime, a significant population of the electronic ground state is expected. Accordingly, the observed intersubband emission is not stimulated but spontaneous. Finally, it is known that the interband radiative rate drops with decreasing temperature due to *p*-type carrier freeze-out. Therefore, we expect a less efficient depopulation of the lowest conduction-band state at reduced temperature; leading to a smaller intersubband signal.

A closer look at the mid-IR signal of the violet LED reveals that the emission around 1000 cm^{-1}, which becomes dominant at 85 K, is mainly due to electron heating. Although it occurs only as a small shoulder on the high-energy side of the blackbody emission, the real intersubband signal

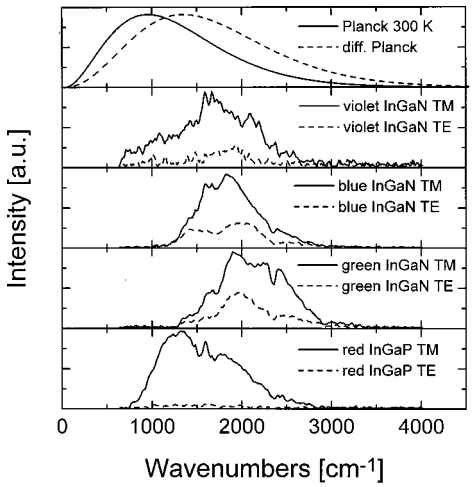

FIG. 3. TM- and TE-polarized electroluminescence spectra of violet, blue, and green InGaN-based, and red InGaP-based LEDs. The bottom figure shows a Planck curve at 300 K and a differential Planck curve for 330/300 K.

is still present at low temperature and has not changed its position. The sharp decrease at the low-energy side of the 85 K violet LED spectrum could be the result of phonon absorption in the material.[15,16] The LO/TO phonon frequencies cited in the literature are 710/741 and 478/694 cm^{-1} for GaN and InN, respectively. Especially for the blue/green LEDs, there are additional absorption features at 1550, 1650, and 2350 cm^{-1}, which are due to water absorption. For the red LED, we finally calculated three bound states in the conduction-band QW. The uppermost state is almost as high as the barrier ground state. The lower transition energy (1064 cm^{-1}) agrees thus quite well with the position of the emission peak at 85 K (1100 cm^{-1}). At room temperature, the signal becomes very broad; this is most likely due to a transition from the continuum into the first excited state and due to electronic blackbody radiation.

In Fig. 3, we present the TM- and TE-polarized mid-IR edge emission spectra of all four LEDs. The InGaN-based devices showed an decreasing TM:TE-polarization ratio with higher-In content. While the violet LED exhibited a polarization ratio of about 3.5:1, this number dropped to 2.75:1 for the blue and to 2.25:1 for the green LED. The decreasing TM polarization, especially of the blue/green InGaN LEDs, is an indication for an intersubband emission originating from quantum boxes rather than QWs. In contrast to this, the red InGaP QW LED exhibited a significantly stronger TM polarization, at a ratio of about 10:1. For intersubband transitions in a QW, the polarization selection rule dictates that the radiation from the edge be purely TM polarized.

One could argue whether the observed signals might be due to transitions into midgap impurity levels. However, such transitions usually lead to an unpolarized, yellowish emission at much higher energies, around 1–2 eV. This is known from the literature, where most of the typical impurities are reported to be located either far from or very close to the band edges. Mg, for instance, which is the shallowest acceptor, is located about 200 meV above the valence-band edge. On the other hand, the nitrogen vacancy, a very common defect in *p*-type GaN, is only 30 meV below the conduction-band edge.[17,18] For *n*-type GaN, Si is the most commonly used dopant; its separation from the conduction-band edge is about 42 meV.[19] Since, in addition, our mid-IR emission depends on the band gap of the InGaN used for the active region, we do not believe we have observed impurity-related radiation.

In order to rule out simple device heating as the cause of the observed mid-IR radiation, we compare in Fig. 3 the room-temperature mid-IR emission spectrum of all LEDs with a Planck spectral distribution at 300 K and a differential Planck curve, which was achieved by subtraction of two Planck curves at 330 and at 300 K. It is obvious that the Planck curves are, in general, broader than our mid-IR emission spectra; in addition, they peak at different energies (1000 and 1200 cm^{-1}).

In conclusion, we have presented a comparison between InGaN/GaN-based LEDs with a different In content of the active region. Mid-IR electroluminescence with peak wavelengths of 1650, 1800, 1950 and 1200 cm^{-1} for the violet, blue, green, and red LEDs were observed. While the mid-IR emission of the red InGaP LED was strongly TM polarized, the InGaN devices showed a decreasing degree of TM polarization with increasing In content. Considering all experimental facts, it is likely that the observed mid-IR electroluminescence has its origin from an active region which contains highly phase-separated InGaN (quantum boxes).

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